

Available for Licensing



Conversion of Methane to Hydrogen and Synthesis Gas Using Bimetallic Oxygen Carriers



Opportunity:

Researchers at the U.S. Department of Energy's National Energy Technology Laboratory (NETL) have developed a novel method for the production of hydrogen (H_2) and synthesis gas (syngas). The process utilizes the chemical looping combustion (CLC) of methane to produce the heat necessary for either methane decomposition to produce pure H_2 or methane reforming to produce syngas. A $CuO-Fe_2O_3$ mixed oxide is used both as an oxygen carrier in the CLC process and as a catalyst (in its reduced state) to produce pure H_2 or synthesis gas via methane decomposition/reforming. This technology is available for licensing and/or further collaborative research.

Overview:

Hydrogen and syngas have many valuable commercial applications including serving as fuels or fuel intermediates. Although both H_2 and syngas can be readily produced from methane, the processes typically generate large quantities of CO_2 , require additional costly processing steps and/or are energy-intensive.

Thus, there exists a need for efficient and cost-effective means of producing valuable H_2 and syngas from methane that also have minimal environmental impact. To this end, NETL researchers have combined Chemical Looping Combustion (CLC) with methane decomposition and reformation to produce H_2 and syngas, respectively. The CLC uses $CuO-Fe_2O_3$ oxide as an oxygen carrier to transport oxygen from air to fuel, avoiding direct contact between fuel and air. Unlike conventional



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combustion, CLC produces a high purity, sequestration-ready CO_2 stream- undiluted by nitrogen (N_2) without expending any major energy required for the separation of CO_2 . This results in a significant cost-savings. Importantly, the CLC process also produces heat that can be used to drive subsequent endothermic chemical reactions such as the conversion of methane into H_2 or syngas. Coupling the CLC of methane with

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methane decomposition and reformation, takes advantage of not only the exothermic nature of CLC, but also the fact that in CLC the bimetallic oxygen carrier cycles between oxidized and reduced states. Significantly, in its reduced state, the $\text{CuO-Fe}_2\text{O}_3$ oxide can function as a catalyst in the methane decomposition and reformation reactions to produce H_2 and syngas, respectively.

A BIAS pelletization method, involving extrusion of a wet paste into pellet shapes, well suited for semi-continuous scale-up has been developed. Pelletization is achieved via the novel combination of inexpensive, readily available fly ash (FA) as a strength additive and low-cost, hydrophobic poly(chloroprene) (PC) as a binder. The combination of FA strength additive and PC imparts both high strength and flexibility.

Significance:

NETL's coupled CLC-methane decomposition/reformation process:

- Produces pure H_2 and syngas and from methane with no CO_2 emissions
- Generates sequestration-ready CO_2 via the CLC process which should contribute considerable cost savings
- Employs a $\text{CuO-Fe}_2\text{O}_3$ CLC oxygen carrier that is highly reactive, stable even after multiple cycles, relatively low cost and environmentally benign
- Uses the reduced oxygen carrier as the catalyst for methane decomposition and reformation to produce H_2 and syngas, respectively
- Requires no external energy as the heat for the methane decomposition/reformation process is supplied by the methane CLC process, making the coupled CLC-methane decomposition/reformation processes highly cost and energy efficient
- Produces a H_2 or syngas stream undiluted by nitrogen, thereby avoiding additional processing steps and associated costs



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Applications:

Any application that involves the conversion of methane to H_2 or synthesis gas. The resulting gasses can be used as feedstocks for conversion to commercial products such as methanol, ammonia, dimethyl ether, and liquid fuels.

Related Patents and Patent Applications:

- U.S. Provisional Patent Application No. **62/265,677** filed December 20, 2015, titled "Production of Pure Hydrogen and Synthesis Gas with Cu-Fe Oxygen Carriers Using Combined Processes of Chemical Looping Combustion and Methane Decomposition/Reforming." Inventors: Ranjani Siriwardane and Hanjing Tian
- U.S. Non-Provisional Patent Application No. **13/159,553** filed June 14, 2011, titled "Regenerable Mixed Copper-Iron-Inert Support Oxygen Carriers for Solid Fuel Chemical Looping Combustion Process." Approved by U.S. patent office, 2016. Inventors: Ranjani Siriwardane and Hanjing Tian.